Nuclear Instruments and Methods in Physics Research A284 (1989) 359–364
North-Holland, Amsterdam

DEPOSITION AND CHARACTERIZATION OF ^{22}Na SOURCES FOR USE WITH MONOENERGETIC POSITRON BEAMS

H. HUOMO *, R. JONES, J. HURST, A. VEHANEN *, J. THROWE **, S.G. USMAR and K.G. LYNN +

Department of Applied Science, Brookhaven National Laboratory, Upton, NY 11973, USA

Received 2 May 1989

We report results on deposition and characterization of sealed 0.5 mCi to 0.25 Ci ^{22}Na β^+ sources. During deposition the homogeneity of the activity distribution of the sources and the yield of energetic β^+ particles was followed in order to optimize the quality.

1. Introduction

During the last ten years studies utilizing moderated and monoenergetic positrons as a matter probe have increased rapidly [1]. The physics of positron moderation and techniques for beam transport are well understood, with the key factor in obtaining higher beam rates being the initial amount of energetic β^+ particles striking the moderator. High intensity monoenergetic positron beams ($> 10^6$ e^+/s) have relied on the use of pair creation from bremsstrahlung generated by energetic electrons from linacs [2,3], or from copper activated in an intense neutron flux [4], or use of short lived ^{58}Co isotope electroplated on a high density holder [5].

Sodium (^{22}Na) is the obvious choice as a source of positrons used with moderate and high intensity beams due to its convenient half-life (2.7 yr) and large branching ratio to positron emission (90%). A serious drawback of ^{22}Na sources is that they are usually fully encapsulated in order to avoid reactions and contamination in the vacuum system of the positron beam. The standard capsules are relatively large, thus prohibiting the use of backscattering moderators [5,6], and the thin window covering the source absorbs a significant fraction of the β^+ particles emitted from the source. The maximum commercially available activity of encapsulated ^{22}Na sources is presently limited to about 150 mCi.

The aim of our study was to investigate the various procedures used in producing high activity sealed and characterized ^{22}Na sources and to fabricate high-activity high β^+ yield sources. An apparatus for the characterization of sealed commercial ^{22}Na sources has been presented by Massoumi et al., but it is not applicable to source characterization during deposition [7].

In this paper we present our source capsule and deposition apparatus designs, the results of tests performed with nonradioactive and radioactive sodium salt solutions, and our methods of source characterization. At the end of the paper the results on our produced sources will be compared to commercially available ^{22}Na sources.

2. Source capsule design

Our source capsule design is shown in fig. 1. It consists of a titanium body with an electron beam welded 5–6 μm thick titanium window. The capsule is closed by press-fit tantalum plug, which is pressed into the window assembly after the source deposition procedure is complete. The source material is deposited directly on the Ti window of the capsule in order to minimize the distance between the moderator and the source, and to easily facilitate the measurement of the source homogeneity and accumulated β^+ and gamma yields. The integrity of the windows was confirmed by pressurizing the window assembly up to 5 atm pressure and subsequently testing for leakage with a helium leak detector. The leak rates of the five tested capsules were below 10^{-8} Torr l/s.

The tantalum plug was designed for maximum backscattering yield to increase the total number of positrons escaping the source. The plug design also incorporates

* Permanent address: Laboratory of Physics, Helsinki University of Physics, 02150 ESPOO, Finland.
** Physics Department.
+ Department of Applied Science and Physics Department.

0168-9002/89/$03.50 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

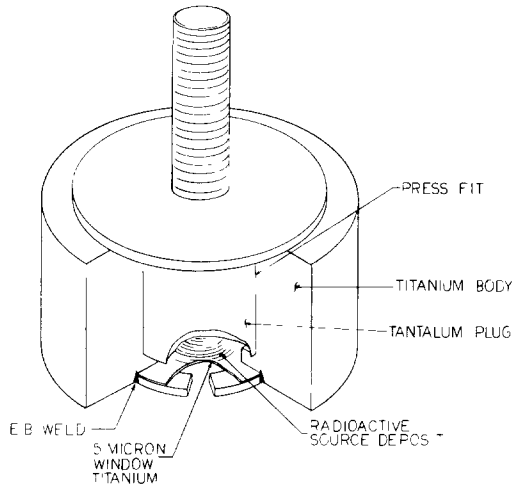

Fig. 1. Source capsule design. The outside diameter of the capsule is 16 mm and the indent above the window has a diameter of 6 mm. The height of the capsule is 10 mm and the assembly can be mounted to the experimental apparatus with the threaded tantalum rod. A long-handled tool was made to mount the source assembly.

free volume for the neon gas produced in the decay of ^{22}Na.

The small indent above the window of the capsule body in fig. 1 allows room for the deposit and any intermediate agents used during deposition.

3. Trial depositions

The key to the production of a useful source is a homogeneous deposit of the sodium salt solution. Numerous tests were made using nonradioactive aqueous sodium chloride and acetate salt solutions. Optical microscopy revealed that aqueous chloride solutions tend to form nonuniform rings of large NaCl crystals. The effect of crystallizing modifiers such as insulin [8], alcohol and 0.5 μm Al_2O_3, as well as various drying rates, indent geometries and surface roughness were investigated. None of these parameters were found to significantly effect the nature in which aqueous chloride solutions dried. Sodium chloride was therefore disregarded as a source material. Also the chloride attack on the thin Ti window was thought to be a potential hazard.

The use of insulin in combination with aqueous acetate solution resulted in a fairly smooth pancake-like deposit. However, radioactive source distribution (see below) produced using aqueous acetate and insulin tended to form an annular shape. A more homogeneous activity distribution was obtained by replacing insulin with an ashless filter paper, as discussed in more detail below.

An aluminum block heater controlled by a variable transformer and monitored by a temperature sensor was constructed to hold the window assembly. In this way the capsule window onto which the solution was deposited could be heated to obtain the most uniform possible deposit. The ashless filter paper was placed on the indent, dramatically increasing the number of nucleation sites for the salt solution. The resulting deposits were quite homogeneous.

The radiation dose to the filter paper is of the order of 100 rad/s (250 mCi source) and thus noticeable deterioration of the filter paper occurs after about one day [9]. As the paper is ash free there is only a small amount of extra residue left in the source. Thus a decrease due to self-absorption is more than compensated by better homogeneity of the deposit.

4. Deposition apparatus

The radioactive ^{22}Na source material is delivered in aqueous solution from the processing company (DuPont NEN *). The concentration of this solution is of the order of 100 mCi/ml in order to reduce radiation damage to the shipping vials and especially to the Teflon coated caps used with the vials. The specific activity of the solution corresponds to 1100–1800 mCi/mg sodium.

Our deposition apparatus is shown in fig. 2 and it consists of a horizontal slide and a 100 μl syringe attached to a vertical slide controlled by a thumb wheel. The source deposition is done in lucite box, lead shielded, which is under small negative pressure. The apparatus exhaust is HEPA filtered.

The ^{22}Na solution (100 mCi/ml) is transferred to the deposition apparatus and brought to dryness in a heated (50 °C) TPX vial. This procedure is repeated until enough ^{22}Na activity exists in the TPX vial for the planned source. The dry ^{22}Na acetate is then brought into solution with 100 μl of Millipore ** water and this solution is drawn to the syringe attached to the vertical slide. Recovery of the sodium from the TPX vial depends strongly on the drying time and temperature, as a too rapid rate leaves a large portion of the sodium acetate attached to the walls of the vial.

The actual deposit on the capsule window is done by adding small drops of about 20 μl of the source and drying for about 30 min at 60 °C. The uniformity of the deposit and the accumulated yield of β^+ and gamma counts was checked after each drop had dried. The ratio of emitted β^+ and gamma counts is sensitive to the

* E.I. DuPont, NEN Products Division, North Billerica, MA 01862, USA.

** Millipore Corp., Bedford, MA 01730, USA.

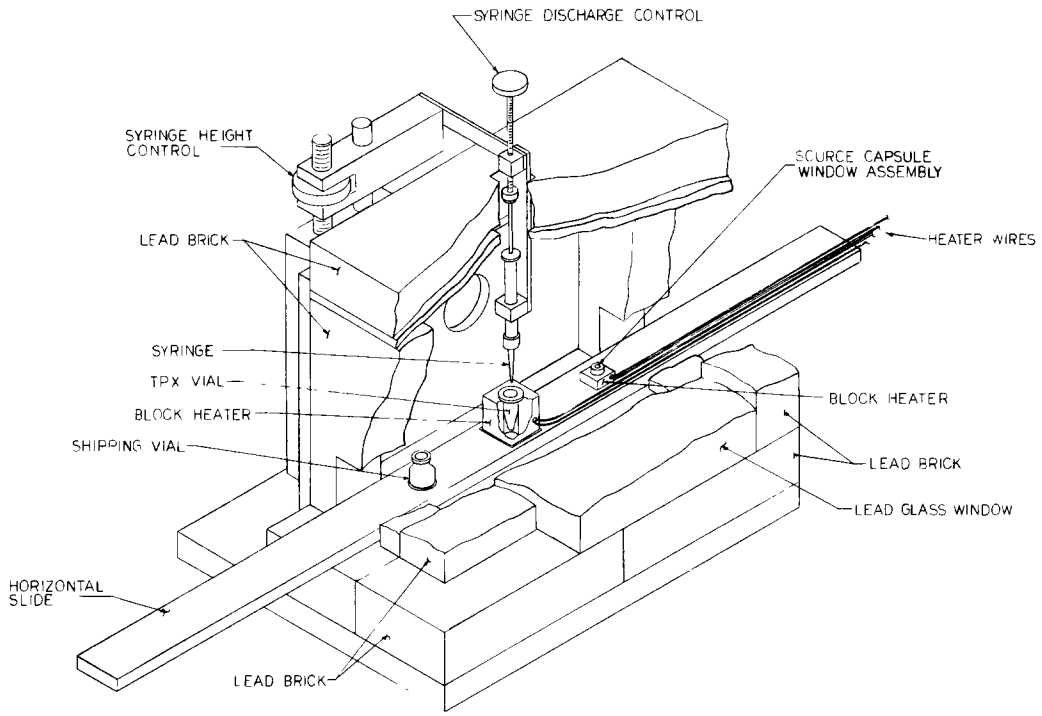

Fig. 2. The source deposition apparatus consists of a horizontal slide and a vertically movable syringe. The shipping vial, the TPX vial and the source window assembly can be moved below the syringe by a long handle (not shown in the figure). The deposition occurs in a ventilated lead housing (thickness 10 cm) with visual observation through a lead-glass window.

uniformity of the deposit. If any islanding or other inhomogeneities occur this ratio decreases.

5. Devices used for ^{22}Na source characterization

Evolution of the spatial distribution of the deposit was followed by observing the light produced by the β^+ particles striking a 1 mm thick cadmium tungstate scintillator placed next to the source. All the β^+ particles are absorbed in the scintillator creating blue-green light. The response to the gamma quanta is only about 3%. The scintillator light was observed visually in the darkened room as shown in fig. 3. The $CdWO_3$ scintillator material was obtained from Harshaw Chemical Co. *

The relative yield of β^+ particles was measured by the counter system shown in fig. 3. The source was placed very close (8 mm) to the scintillator coupled to a photomultiplier tube. Separation of β^+ and gamma counts was obtained by the use of a movable shutter (Al, thickness 3 mm) between the source and the scintillator (see fig. 3).

Calibration of this counting system for both β^+ and gamma response was obtained by direct comparison using the unsealed sources reported in a previous study of moderators [10].

* Harshaw Chemical Co., Solon, OH 44139, USA.

6. Results of the ^{22}Na source production

We produced three sources with the techniques described above. Source strengths in chronological order were 15 mCi, 70 mCi and 250 mCi. Total hand radiation dose from these operations was about 3 rem and body dose about 0.6 rem for the two persons involved in the deposition of the sources. These trial sources included a large amount of extra handling of the source material and the source itself during initial experimental setup.

Fig. 4 shows the evolution of the β^+ counts as a function of gamma counts for consecutive drops added to the source. The data in fig. 4 for all sources shows the same relation between the evolution of the β^+ and gamma counts. During the production of the 15 mCi source one drop landed accidentally on the ledge of the source capsule indent. This can be seen as an abrupt horizontal shift in the plot, with the gamma yield increasing while the β^+ yield remains almost constant. The last drop was dried very slowly at about 30 °C for 20 h resulting in an increased β^+ yield (see fig. 4, left panel). An additional gain in the β^+ yield ($\approx$ 15%) was

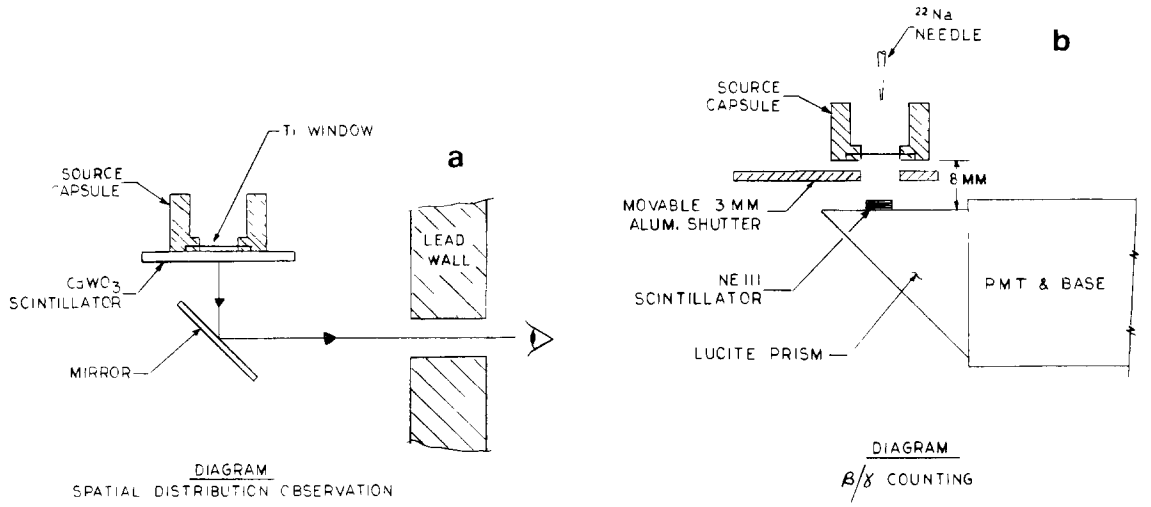

Fig 3. Source characterization apparatus consists of a source homogeneity observation station and a counting station. The source homogeneity is observed by a $CdWO_3$ scintillator placed next to the source window, and the scintillation light from the β^+ particles is observed visually in a darkened room. In the counting setup the difference between β^+ and gamma response is separated by a movable aluminum shutter between the source and the thin scintillator.

obtained by the insertion of the press-fit tantalum plug. We note that the gamma yield also increased due to increased backscattering.

For the 70 mCi source (fig. 4, middle panel) we had to refill the TPX vial with the shipping solution to obtain more radioactive material. The heating rate was obviously too high during the volume reduction, and a major fraction of the sodium salt became attached to the walls of the TPX vial. This resulted in a very low specific activity after addition of 100 μl of water. The sodium salt was recovered by filling the TPX vial and evaporating the water at a lower temperature. Increase in the β^+ yield ($\approx 8\%$) as in the case of the 15 mCi source was also observed here, as the tantalum plug was press-fit to the capsule body.

Deposition of the 250 mCi source was made from the highest specific activity sodium acetate solution (see fig. 4, right panel). After the fifth drop we had to add more shipping solution to the TPX vial and heat it slowly to dryness. This setup required about two days and during that time the ashless filter paper was radiation damaged. The color of the paper had changed and the spatial distribution of the deposited source was very inhomogeneous. The radiation damage to the paper had reduced the number of available nucleation sites and as a result the activity contained 15–20 small hot spots.

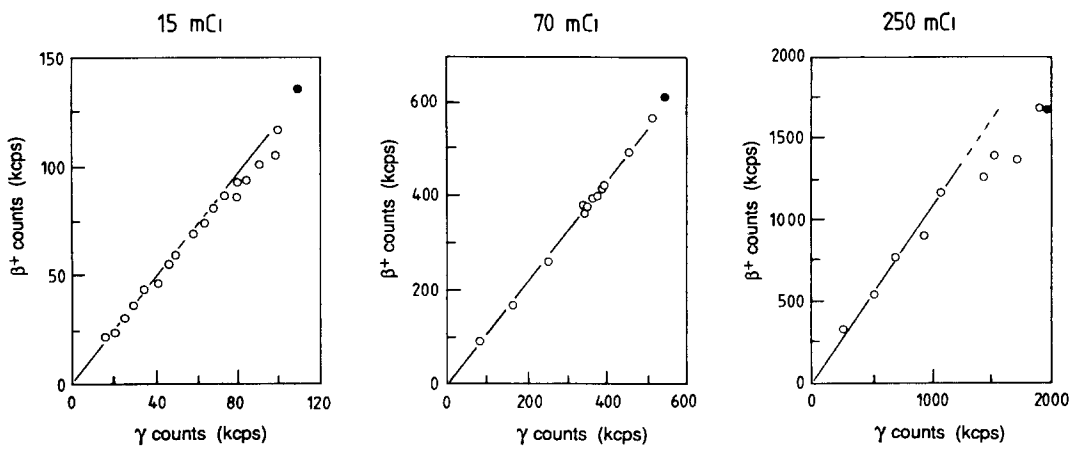

Fig. 4. The β^+ response of the ^{22}Na sources during deposition is shown as a function of the gamma-ray response using the counting system shown in fig. 3. The final measurement for the closed source assembly is marked with a closed symbol.

Table 1
Total activity and the yield of β^+ particles from several sources

Source (fig. 5)	Source ID	Active area diameter [mm]	Source capsule diameter [mm]	Source activity [mCi]	β^+ emission [%]
	BNL -87				
1	BNL #A	4	16	0.5	24 [a]
2	BNL #B	4	16	130	6 [a]
	BNL -88				
3	BNL #C	6	16	15	25
4	BNL #D	6	16	250	16
5	BNL #E	6	16	70	21
	(NEN DuPont)				
6	NEN #1	4	13	140	26
7	NEN #2	3	13	100	14
8	NEN #3 (donut)	6×4 (diam.×h)	16	10	18

[a] Sources prepared earlier under less controlled conditions (see text).

These hot spots covered about 50% of the total source area and they were evenly distributed. This islanding of the source was also observed in the resulting β^+ yield as a change of slope for the next three drops added to the source. The last drop was dried again very slowly to increase the homogeneity. In this case the closure of the capsule with the tantalum plug made a negligible change to the β^+ yield.

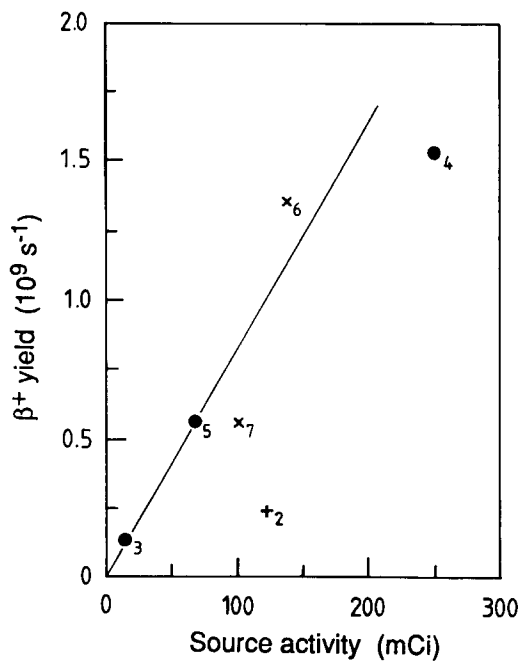

Fig. 5. Final β^+ and gamma yields of the ^{22}Na sources are shown (○), together with two commercially obtained (DuPont NEN) ^{22}Na sources (×). The labels of the sources are described in table 1. The solid line in the figure represents the maximum possible β^+ yield from the current capsule design with the usage of the ashless filter paper as an intermediate agent. An earlier source made under initial, less-controlled setup is also shown (+).

Fig. 5 shows the source activity of several sources produced, together with two commercial sources of similar design. Table 1 contains more specific information on the sources. It should be noted that the BNL sources A and B (see table 1) were produced earlier by remote manipulation inside a hot cell. Insulin was used as a dispersing agent, heating was uncontrolled, and β^+ yield was not monitored during deposition. Occasionally, a fraction of active source material was lost and the resulting deposition was inhomogeneous. The poor quality of the resulting sources (see fig. 5) motivated the development of the techniques presented in this paper. The solid line in fig. 5 represents the maximum possible ratio between the source activity and the β^+ yield which we could obtain, for the current source capsule design, including the use of ashless filter paper as a intermediate agent to increase the source homogeneity.

All the sources described in this paper are currently in use for slow positron beam generation. Leak rates through the press-fit capsules are observed to be compatible with UHV requirements. The sources are bakeable to at least 100 °C. Also the rigidity of the deposit has been strong enough to permit repeat handling of the source in any installation geometry without mechanical changes in the deposit. No degradation of operation of the relative β^+ yield has been observed over a period of ~ 1 year.

During the whole manufacturing process of the good-quality sources less than 5% of the initial ^{22}Na activity was lost, mainly as a residue in the drying vial and in the shipping vial. Small amounts of sodium that become airborne during the drying of the shipping solution leave contamination in the vicinity of the top of the drying vial. No contamination was found on the

inside surfaces of the source preparation apparatus nor in the closed HEPA filter assembly.

7. Conclusion

In this report we have presented methods to produce well-characterized sealed ^{22}Na sources for use in slow positron beam generation. The beta–gamma counting system and the $CdWO_3$ scintillator have provided a new understanding of the deposition process as well as a means of evaluating completed sources. The following guidelines for source deposition were determined:

(1) The rate of drying of the source solution in the TPX vial and in the source capsule must be carefully controlled to prevent loss of material and to promote uniformity of deposition.

(2) Ashless filter paper can be used to provide nucleation sites for uniform deposition. For high-activity sources, deposition should be completed as rapidly as possible so that the filter paper does not have time to deteriorate. A relatively long drying time after each drop of deposition should be allowed.

We also emphasize that the net intensity of various ^{22}Na-based slow positron beams is strongly affected by the initial β^+ yield of the radioactive source as it can vary significantly, as seen in both in our sources and in the commercial ones. (See also ref. [7].) Further improvements in the β^+ yield may be obtained by using a thin beryllium window instead of titanium.

Acknowledgements

The authors would like to thank C. Marchetti and other staff members at DuPont NEN for useful discussions and L. Mountjoy and E. Gramsch for technical assistance. Mr. J. Dioguardi's contributions in apparatus design, construction coordination, and art work was invaluable. Work supported by US Dept. of Energy, Division of Materials Sciences, Office of Basic Energy Sciences under Contract no. DE-AC02-76CH00016 and the Academy of Finland.

References

[1] P.J. Schultz and K.G. Lynn, Rev. Mod Phys. 60 (1988) 701.
[2] R Howell, R.A. Alvarez and M. Stanke, Appl. Phys. Lett 40 (1982) 751
[3] R. Ley, K.D Niebling, A Osipowicz, A. Piccard and G. Werth, in: Positron Annihilation, eds. P.C. Jain, R M. Singuru and K.P Gopinathan (World Scientific, Singapore, 1985) p. 996.
[4] K G. Lynn, A.P. Mills, Jr., R.N. West, S. Berko, K.F. Canter and L.O. Roellig, Phys. Rev. Lett. 54 (1985) 1702.
[5] J. Lahtinen, A Vehanen, H. Huomo, J. Makinen, P. Huttunen, K Rytsölä, M Bentzon and P. Hautojärvi, Nucl. Instr. and Meth. B17 (1986) 73.
[6] A. Vehanen, K.G. Lynn, P.J. Schultz and M. Eldrup, Appl. Phys. A32 (1983) 163.
[7] G.R. Massoumi, P.J. Schultz, W.N. Lennard and J. Ociera, Nucl. Instr. and Meth B30 (1988) 592.
[8] L.M. Langer, Rev. Sci. Instr. 20 (1949) 216
[9] In Radiation Effects on Organic Materials, eds. R.O. Bolt and J.G Carroll (Academic Press, New York, 1963).
[10] E. Gramsch, J Throwe and K.G. Lynn, Appl Phys. Lett. 51 (1987) 1862.